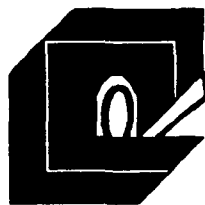


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COHERENT COOPERATIVE RADIATION IN SOLIDS



COLLEGE OF ENGINEERING

DEPARTMENT OF ELECTRICAL ENGINEERING & COMPUTER SCIENCE

UNIVERSITY OF MICHIGAN, ANN ARBOR, MICHIGAN

FINAL TECHNICAL REPORT

Contract F49620-88-C-0079

Principal Investigator: Professor S.C. Rand

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19. Abstract

The principal objective of this research has been to characterize cooperative transitions of coupled, dopant ions in solids and to investigate potential applications for novel lasers. Major accomplishments of the program after three years include demonstration of the first continuous-wave lasers pumped by pair and trio transitions, development of a cooperative upconversion laser and a description of its unusual dynamics. These Er^{3+} lasers were pumped optically at $1.5\ \mu\text{m}$ and operated at $2.8\ \mu\text{m}$ and $0.85\ \mu\text{m}$ by virtue of cooperative pair and trio excitation processes respectively. The cw trio laser achieved an efficiency of nearly 30% on the $^4\text{S}_{3/2} - ^4\text{I}_{13/2}$ transition which is "self-terminating" at low intensities, a surprisingly high value. This efficiency is the highest of any upconversion laser reported to date and illustrates the important potential of this excitation mechanism for sustaining inversions on new transitions in rare earth lasers.

Avalanche upconversion dynamics and associated nonlinear optical response was also reported for the first time in $\text{Tm}:\text{YALO}$ crystals at room temperature. The avalanche mechanism involves a cooperative down-conversion (cross relaxation) step and is a promising method of pumping rare earth lasers at entirely new (diode compatible) wavelengths, corresponding to excited state absorptions rather than ground state absorptions. Quantum theory describing the threshold for photo-darkening and the role of energy migration in avalanches was developed, and semi-quantitative agreement obtained with preliminary measurements of nonlinear susceptibilities. Cooperative dynamics in general, and avalanche dynamics in particular, were shown to furnish a promising new mechanism not only for solid state lasers but for nonlinear optical interactions as well, opening the door to practical signal processing and switching applications exploiting the low intensity thresholds of avalanche processes.

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SECTION 1

SUMMARY OF RESEARCH ACCOMPLISHMENTS

This section presents a brief description of key advances made during the contract period 5/1/88-10/31/91. Section 2 presents a list of published papers, and conference and colloquium presentations made during the report period. Patent applications filed within the report period are listed in Section 3. More detailed descriptions of the scientific work itself, as well as its implications and applications are described in reprints of scientific papers published during the contract period, and included as an appendix in this report.

LIST OF MAJOR ACCOMPLISHMENTS

1. Demonstration of the first cw pair-pumped solid state laser. Operation at room temperature. See Section 3 on patents.
2. *Demonstration of the first cw trio-pumped solid state laser. Operation with very high efficiency at liquid nitrogen temperature. See Section 3 on patents.*
3. Rate equation analysis achieves basic consistency with the observation of continuous inversion sustained on a normally self-terminating transition with lower laser level pumping.
4. Development of quantum theory from the Liouville equation capable of describing near neighbor coherent dynamics as well as energy migration over macroscopic distances.
5. Prediction and preliminary observation of sustained, oscillatory output from pair-pumped lasers at threshold and well above threshold.
6. Discovery of avalanche upconversion at room temperature in Tm:YALO and microscopic explanation of the origin of threshold behavior.
7. Demonstration of two-beam coupling (nonlinear mixing) mediated by cooperative nonlinearities.
8. Discovery of spatial modulation of laser beams by cooperative nonlinearities (far field ring patterns).

SECTION 2

PUBLICATIONS & PRESENTATIONS

This section presents a list of publications, as well as conference and colloquium presentations made during the contract period (May 1, 1988 - October 31, 1991). Reprints of substantial scientific papers are attached as appendices to this report.

LIST OF PUBLICATIONS AND PRESENTATIONS

1. (Invited contribution) S.A. Pollack, D.B. Chang, M. Birnbaum and S.C. Rand, Upconversion Lasers Excited by Pairs and Trios, Optics News 14, No.12, 21(1988).
2. (Invited) S.C. Rand, Pair Processes in Coherent Optical Interactions, XVI International Quantum Electronics Conference, 18-21 July 1988, Tokyo, Japan.
3. (Invited) S.C. Rand, Pair-pumped Upconversion Solid State Lasers, Laser Materials and Laser Spectroscopy, 25-27 July 1988, Shanghai, PRC.
4. (Invited dept. colloquium) S.C. Rand, Pair Processes in Coherent Optical Interactions, Dept. of Physics and Astronomy, University of Toledo, Toledo, Ohio, September 29, 1988.
5. (Invited Dept. colloquium) S.C. Rand, Pair Processes in Coherent Optical Interactions, Dept. of Physics and Astronomy, Oakland University , Oakland, Michigan, October 12, 1988.
6. S.C. Rand, Pair Processes in Coherent Optical Interactions, Harry Diamond Laboratories, Germantown, Maryland, February 2, 1989.
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8. S.C. Rand, Collisions Gelees, Colloque du departement d'optique, Universite de Lyon, 1989.
9. P. Xie and S.C. Rand, Pair-pumped Continuous-wave Solid State Laser, in Digest of Topical Meeting on Advanced Solid State Lasers, 1990 (Optical Society of America, Washington, DC.) Salt Lake City, Utah, March 5-7, 1990, pp.179-181.
10. P. Xie and S.C. Rand, Pair-pumped, Continuous-wave Upconversion Laser, in Digest of Conference on Lasers and Electro-optics 1990, IEEE/Leos and Optical Society of America, Anaheim, California, May 21-25, 1990, paper CME4.
11. P. Xie and S.C. Rand, Trio Upconversion Laser, Annual Meeting of the Optical Society of America, Boston, Massachusetts, November 4-9, 1990, paper THL8.
12. P. Xie and S.C. Rand, Continuous-wave Pair-pumped Laser, Optics Letters 15, 848(1990).
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14. S. Rand, Upconversion Lasers and coupled Atom Physics, Graduate seminar in Modern Physics, Dept. of Physics, University of Michigan, 1990.
15. S. Rand, Cooperative Nonlinear Optics, Applied Physics Seminar, University of Michigan, January 30, 1991.
16. S.C. Rand (invited), Cooperative Optical Nonlinearities, Quantum Electronics and Laser Science Conference (QELS '91), Baltimore, MD, May 12-17, 1991, paper QTHK1.
17. D. Redman (invited), S. Brown, S. Rand and S. Satoh, High Resolution Four-Wave Mixing Spectroscopy of Nitrogen-Vacancy Centers in Diamond, Quantum Electronics and Laser Science Conference (QELS'91), Baltimore, MD, May 12-17, 1991, paper QMA7.

18. P. Xie and S.C. Rand, Cooperative Upconversion Lasers, Conference on Lasers and Electro-optics (CLEO'91), Baltimore, MD, May 12-17, 1991, paper CTuW20.
19. Q. Shu and S.C. Rand, Beam Coupling by Cooperative Nonlinearities, Conference on Lasers and Electro-optics (CLEO'91), Baltimore, MD, May 12-17, 1991, paper CWL5.
20. P. Xie and S.C. Rand, Continuous-wave, Pair-pumped Laser, in Advanced Solid State Lasers, Vol. 6, eds. H.P. Jenssen and G. Dube, Optical Society of America, 1991, pp. 190-191.
21. H. Ni and S.C. Rand, Avalanche Upconversion in Tm:YALO, Optics Letters 16, 1424(1991).

SECTION 3

PATENT APPLICATIONS

1. S.C. Rand, Continuous-wave, Pair-pumped Laser, Patent Application Serial No. 487,885 filed March 2, 1990 (Assignment to University of Michigan). Under review.
2. S.C. Rand, Trio Upconversion Laser, Continuation-in-Part (CIP) filed May 16, 1990 for Patent Application Serial No. 487,885 filed March 2, 1990 (Assignment to University of Michigan). Patent issued August 6, 1991 (No. 5,038,358).

APPENDIX A
Reprints of Published Papers



XVI INTERNATIONAL CONFERENCE

ON

QUANTUM ELECTRONICS

18-21 JULY 1988

TOKYO, JAPAN

Digest of Technical Papers

Organized by

Japan Society of Applied Physics

Co-Sponsored by

Institute of Electrical Engineers of Japan

Institute of Electronics, Information and

Communication Engineers

Laser Society of Japan

International Union of Pure and

Applied Physics (IUPAP)

Optoelectronic Industry and Technology

Development Association (OITDA)

Cooperative Support by

International Commission for Optics (ICO)

American Physical Society (APS)

Lasers and Electro-Optics Society of

the Institute of Electrical and Electronics Engineers

Optical Society of America

Quantum Electronics Division of

the European Physical Society

PAIR PROCESSES IN COHERENT OPTICAL INTERACTIONS

Stephen C. Rand, Depts. of Physics and Electrical Engineering,
University of Michigan, Ann Arbor, MI 48109.

Recent work indicates that an interesting class of nonlinear optical phenomena exists in which weak electronic coupling between clusters of a few atoms has an important influence on coherent optical interactions. In this paper we consider primarily pair-pumped upconversion lasing, optical suppression of the Van der Waal's force, seven-line resonance fluorescence spectra in stationary pair systems, two-atom coherence and dark processes in four-wave mixing spectroscopy.

Cooperative effects in the radiation from small groups of atoms develop as the result of short-range multipole-multipole or exchange interactions.

They occur in gases, liquids and solids and are quite different from effects like superradiance in which interatomic coupling is furnished by the radiation field itself or multi-photon processes in which a sequence of linear processes or the nonlinear response of single atoms is involved. Simple electric dipole-dipole coupling between atoms combined with the driving force of strong coherent light fields is enough to give rise to a rich variety of new phenomena.

Pair interactions can have significant impact on the operation of lasers even when energy downconversion is involved. For example the Tm,Ho:YSGG

and Tm:YSAG lasers have quantum efficiencies close to two because a single incident photon can result in two upper laser state ions [1] as the result of pair processes. However, here we are concerned mainly with pair interactions which furnish energy upconversion, particularly in solid state laser materials. Frequency upconversion of radiation and even laser pumping may be readily accomplished by multiphoton processes relying on linear or nonlinear susceptibilities of individual atoms. However, extremely efficient upconversion can also occur by cooperative multi-atom processes involving simultaneous transitions on two or more atoms.

Cooperative pair pumping in 5% Er:YLF has recently been shown [2] to produce laser action from upconverted energy states following pumping at 1.54 microns with an Er-glass laser on the ground to $^4I_{13/2}$ transition.

Two $^4I_{13/2}$ ions decay cooperatively to yield one $^4I_{9/2}$ ion, competing effectively with spontaneous emission because of strong inter-atom coupling and long $^4I_{13/2}$ lifetime. The experiments in Er:YLF have shown that inversion of $^4I_{11/2}$ with respect to lower states is achieved by the multi-atom upconversion mechanism alone and that cw operation is possible in this new class of laser pumped by energy "pooling".

On a fundamental level it has also been found that the problem of optical transitions of two coupled atoms can be calculated exactly for strong fields using dressed atom techniques. For stationary or nearly stationary atoms, the resonance fluorescence spectrum and the stimulated emission spectrum versus detuning can be obtained in this fashion and reveal

unexpected features. The resonance fluorescence spectrum is predicted to consist of seven lines rather than the usual three because transitions occur between triplets of dressed states rather than doublets [3]. Also, coherent dips appear in the stimulated emission spectrum when the Rabi frequency reaches a value determined by the strength of the interatomic coupling [4]. These features are due to a dynamic decoupling of the atomic pair interactions called the electric magic angle effect. At the magic angle condition the Van der Waal's force between the coupled atoms is suppressed.

Experimental results from four-wave mixing spectroscopy have also revealed that, with nearly degenerate input frequencies and counter-propagating pump waves, the phase conjugate output intensity is sensitive to dark processes involving pairwise cross-relaxation of ions in crystals [5]. In Beta-Alumina doped with high concentrations of trivalent Nd, weak pair processes obscured in conventional fluorescence studies by overwhelming impurity relaxation have been observed. Lineshape analysis of the (1 kHz wide) four-wave mixing spectrum reveals the onset of cross-relaxation at 6×10^{20} Nd/cm³, whereas fluorescence decay times are constant throughout this doping range. Hence pair processes can have a significant effect on third order nonlinear optical susceptibilities. It is to be expected that in new crystal systems where the majority of absorbers consist of pairs, novel aspects of de-localized coherences induced on two spatially separate atoms will be encountered.

This work was supported by AFOSR.

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TUESDAY 19 JULY 1988

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(A SATELLITE MEETING OF IQEC '88)

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July 25-27, 1988

Editors

Wang Zhijiang

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Optics, Fudan University, China*



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PAIR-PUMPED UPCONVERSION SOLID STATE LASERS

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University of Michigan, Ann Arbor, MI 48109.

Recent work has revealed an interesting class of nonlinear optical phenomena in which weak, electronic coupling between clusters of a few atoms has an important influence on coherent optical interactions. In this paper we consider primarily the observation of pair-pumped upconversion laser action in rare-earth-doped solids. However we also describe briefly several other new effects which are similarly due to multi-atom processes. These include optical suppression of the Van der Waal's force, the occurrence of seven-line resonance fluorescence spectra in stationary pair systems, and the influence of dark pair processes in four-wave mixing spectroscopy.

Cooperative effects in the radiation from small groups of atoms develop as the result of short-range multipole-multipole or exchange interactions. They are quite different from effects like superradiance in which interatomic coupling is furnished by the radiation field itself or multi-photon processes in which sequential linear processes or nonlinear response of single atoms is involved. The mechanism for the results reported here depends on simple electric dipole-dipole coupling between atoms combined with the driving force of intense coherent light.

In the past, pair interactions had significant impact only on the operation of lasers in which energy downconversion played a role. For example, the N-line ruby laser operated on a difference pair transition near 703 nm instead of the usual 694 nm transition. The more recent Tm:Ho:YSGG and Tm:YAG lasers have quantum efficiencies close to two, rather than unity, because single incident photons are able to promote two ions to the upper laser level via nearly resonant energy transfer processes involving dopant pairs. However, we are principally concerned here with pair interactions in solids which furnish energy upconversion through energy "pooling". Of course multiphoton processes relying on linear or nonlinear susceptibilities of individual atoms are commonly used for frequency upconversion of radiation and even laser pumping. However, extremely efficient upconversion can also occur by cooperative multi-atom processes involving simultaneous transitions on two or more atoms.

Cooperative pair pumping in 5% Er:YLF has recently been shown to produce laser action from an upconverted energy state following pumping at 1.54 microns with an Er:glass laser on the ground state ($J=15/2$) to first resonance ($J=13/2$) transition. In Er crystals with low rare earth concentrations, the lifetime of the second excited state ($J=11/2$) normally exceeds that of $J=13/2$ and laser action on the 11/2 to 13/2 transition is quenched. However, at high Er concentrations pair interactions contribute to depletion of the lower level and laser action becomes possible. In the Er system, two 13/2 ions decay cooperatively to yield one high energy ion in the 9/2 state, which relaxes rapidly to the upper laser level ($J=11/2$). The cooperative process has a very high rate

because of strong inter-atom coupling and the long $13/2$ lifetime in YLF. Its rate greatly exceeds the spontaneous decay from the same state as well as the filling rate from higher states at modest input powers. The upconversion process is shown for the first time to be so effective in Er:YLF that this mechanism alone can invert $11/2$ with respect to lower states. Cw operation is also predicted, despite the fact that atoms are excited directly to the lower laser level by pump radiation. The interatomic coupling merely has to be strong enough so that depletion exceeds the filling rate in the lower level. Analysis of saturation and coherence effects in laser systems of this kind will be presented.

Other effects of pair interactions on coherent optical phenomena which have been observed or calculated recently are also reported. The pair resonance fluorescence spectrum for example is predicted to consist of seven lines, rather than the usual three, because transitions occur between triplets of dressed states rather than doublets. Also, coherent dips are predicted in the stimulated pair emission spectrum when the Rabi frequency reaches a value determined by the strength of the interatomic coupling. These coherent features are due to dynamic decoupling of the atomic pair interactions by the light field, a phenomenon called the electric magic angle effect. At the magic angle condition, the Van der Waal's force between the coupled atoms is suppressed.

In addition, experimental results from nearly degenerate four-wave mixing spectroscopy (NDFWM) have revealed that the coherent signal

intensity is sensitive to non-radiative (dark) processes involving cross-relaxation of ionic pairs in crystals. In the promising laser host material Beta"-Alumina, weak pair processes are observed at high concentrations of trivalent Nd dopant ions with NDFWM, but are obscured in conventional fluorescence studies by overwhelming decay contributions from other impurities. Lineshape analysis of the (1 kHz wide) four-wave mixing spectrum reveals the onset of cross-relaxation at 6×10^{20} Nd/cm³, although excited state decay times measured in fluorescence are constant throughout this doping range. Saturation intensities determined from the NDFWM results also decrease by an order of magnitude in this density range, establishing that the maximum useful density of Nd ions in this material for laser applications is limited by a dark pair process.

Hence pair interactions can have major effects on practical laser operation and in future will permit construction of efficient lasers with operating wavelengths much shorter than the excitation wavelengths. It is now known that pair processes can be enhanced in special crystal systems where the majority of absorbers consist of pairs or trios of atoms. In such solids it is to be expected that higher upconversion efficiencies and further novel aspects of such interactions will be encountered in studies of stimulated emission and de-localized coherences induced on spatially separated atoms.

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Continuous-Wave, Pair-Pumped Laser

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Abstract

We report room temperature operation of the first continuous-wave laser which relies exclusively on cooperative upconversion by coupled ionic pairs to achieve population inversion.

Experiment

The energy -level diagram of the Er^{3+} ion is shown in Fig. 1. The crosses on energy level $^4I_{13/2}$ represent erbium ions resonantly excited with a cw color center laser operating at $1.51\mu\text{m}$. Ion-ion interactions induce cooperative energy upconversion as shown in the figure. The curved arrows indicate the dynamics schematically, with ion 2 losing energy by returning to ground state, and ion 1 gaining energy by upconversion to the energy level $^4I_{9/2}$ (lifetime $\sim 7\mu\text{s}$) [4] which then relaxes rapidly to populate the upper laser level $^4I_{11/2}$. In dilute $\text{Er}:\text{CaF}_2$ crystals the transition $^4I_{11/2} - ^4I_{13/2}$ is self-quenched because the $^4I_{13/2}$ lifetime exceeds that of $^4I_{11/2}$ by at least a factor of two [2]. However $\text{Er} - \text{Er}$ interactions in concentrated crystals make the $^4I_{13/2}$ lifetime excitation dependent. As a result even when the lower laser level is pumped, as in the present work, it becomes possible to achieve steady-state inversion from upconverted states without the assistance of multiphoton excitation. This agrees with an exact rate equation analysis of the dynamics which includes 2-photon absorption to $^4I_{9/2}$ [5] and a stability analysis [5] which uses the Routh - Hurwitz criterion and shows the absence of sustained relaxation oscillations in the system.

Cw Er^{3+} laser output power at $2.75\mu\text{m}$ versus absorbed input pump power at $1.51\mu\text{m}$ is shown in Fig. 2. The upconversion laser operated in the fundamental TEM_{00} mode. With 2% output coupling it generated a maximum cw output power of 9 mW with 200mW pump power (4.5% overall efficiency)

from an NaCl color center laser. The observed threshold was 105 mW. The evidence that laser emission is due exclusively to the upconversion mechanism and not to two-photon absorption is shown in Fig. 3. In this figure the time dependence of $^4I_{11/2}$ population is monitored by detection of 985nm fluorescence following short pulse excitation. No prompt fluorescence peak was observed within the excitation pulsewidth. Instead, a slow risetime is observed after termination of the pump pulse, clearly indicating that upconversion proceeds purely by pair spontaneous processes in the long-lived $^4I_{13/2}$ state at the low excitation intensities in this experiment.

Research sponsored by the Air Force Office of Scientific Research (AFSC) under Contract F49620-88-C-0079.

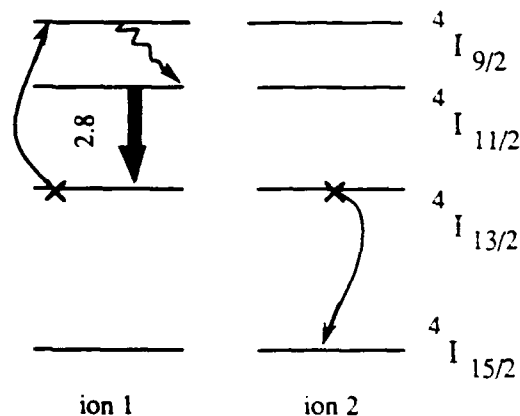


Figure 1. Energy levels involved in the cw pair-pumped erbium laser. The curved arrows indicate the cooperative pair process involving two resonantly excited ions which results in population inversion in the upconverted energy state $^4I_{11/2}$.

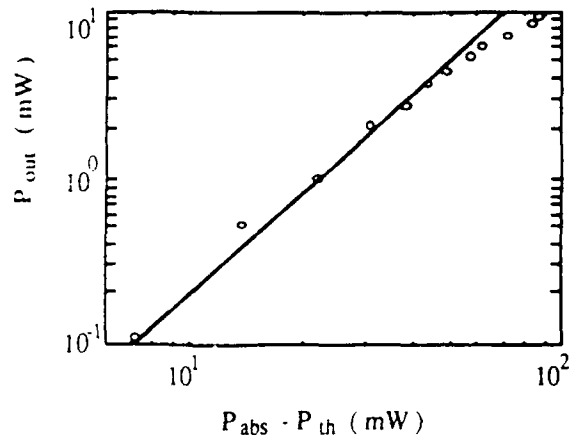


Figure 2. Log - Log plot of cw output power of the $\text{Er}^{3+}:\text{CaF}_2$ upconversion laser vs absorbed cw input power above threshold. The solid curve illustrates the quadratic dependence followed at low powers and shows the onset of saturation at higher powers.

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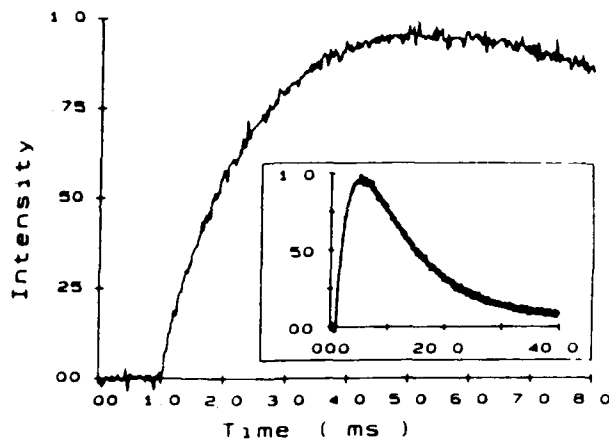


Figure 3. $4I_{11/2} - 4I_{15/2}$ fluorescence versus time at 985nm excited by a $30\ \mu\text{s}$ (acousto-optically tailored) rectangular laser pulse at $\lambda = 1.51\ \mu\text{m}$, also shown in the figure. The absence of fluorescence response during the excitation pulse indicates that the upper laser level is only populated after termination of pumping. This upconversion therefore occurs entirely by the pair process indicated in Fig. 1 (2ms / div.).

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QMA7 Ultrahigh resolution four-wave-mixing spectroscopy of nitrogen-vacancy centers in diamond

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The nitrogen-vacancy center in diamond exhibits a zero-phonon transition at 637 nm that has been assigned as an $A \rightarrow E$ transition at a site with trigonal symmetry.¹ Paramagnetic resonance studies revealed a triplet feature that was originally assigned to an excited state.² More recently, it was argued that the triplet signal actually arose from the ground state.^{3,4} We employ a new method of ultra-high-resolution nearly degenerate four-wave-mixing (NDFWM) spectroscopy, as well as persistent hole burning and electron paramagnetic resonance, to measure the metastable spin singlet excited-state lifetime and the ground-state spin-lattice relaxation time and also to prove that the ground state is indeed a triplet.

The NDFWM experimental arrangement is shown in Fig. 1. The single-mode ring dye laser is tuned to the zero-phonon transition at 637 nm at a temperature of 80 K. The effects of laser jitter are eliminated by deriving all beams from the same highly stabilized laser. The probe beam is upshifted by the first A-O modulator frequency and is subsequently downshifted by the second. The probe is detuned by stepping the frequency of the second synthesizer for coarse resolution or, in a novel development, by imposing a linear phase ramp on the rf signal to the second A-O modulator for subhertz resolution. This improvement increases resolution by nearly three orders of magnitude over previous work and is limited only by the stability of the phase driving electronics and the instantaneous linewidth of the laser. Synchronous detection is employed throughout; however, the narrowness of the resonance in the high-resolution scan prevents chopping of an input beam in order to avoid modulation sidebands in the spectrum. The wide frequency scan employs a new development in which the pump and probe beams

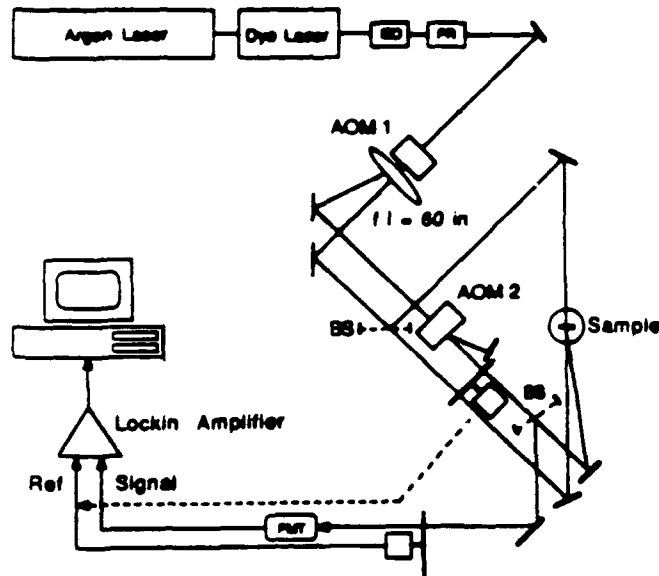
are chopped at different frequencies and the signal is detected at the sum frequency. This intermodulation technique eliminates dc background and improves the signal-to-noise ratio by a factor of 20.

Figure 2 shows the signal-averaged NDFWM spectrum for two different frequency sweep ranges at a temperature of 80 K. The 10-Hz scan shows an ultranarrow resonance with a FWHM of 1.8 Hz. Each point represents a step of 200 mHz. The inset shows a wide scan exhibiting a broad peak with a width of 372 Hz. The two resonances are indicative of two decay times in the optical excitation. The theory of NDFWM predicts a peak centered at zero detuning for each ground state decay channel associated with the system, each having a FWHM proportional to the decay rate. The triplet-triplet zero-phonon transition is strongly allowed with a fluorescence decay rate of 13 ns,⁵ so the ultranarrow resonance must arise from a metastable $S = 0$ excited state with a spin-forbidden transition lifetime of 555 ms. The broader peak corresponds to a decay time T_1 of 2.7 ms, consistent with the spin-lattice decay time measured in ESR experiments, which we describe next.

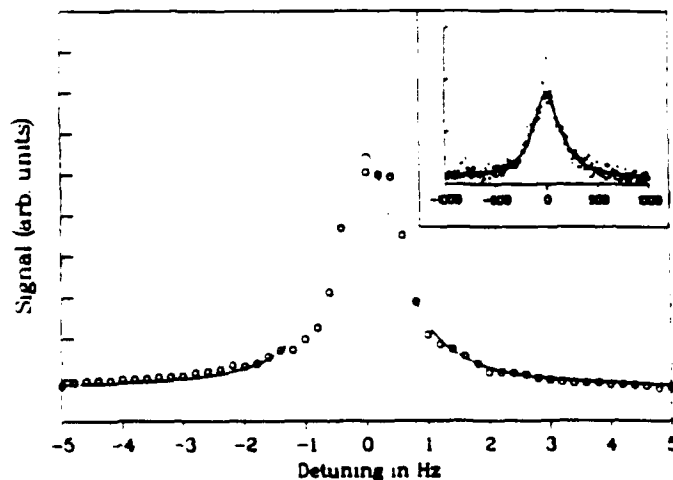
To prove that the triplet signal arises from the ground state, paramagnetic resonance spectra were recorded at a frequency of 9.2 GHz with no optical excitation. Exceptionally low saturation power of the N-V microwave resonances required powers below 2 μ W. The signal strength followed a Curie law temperature dependence, indicative of an isolated ground state. The rate was independent of both concentration and optical power, indicating that the decay resulted from spin-lattice relaxation. Furthermore, T_1 was determined to be 3.3 ms at room temperature by measuring the decay of the signal upon removal of optical excitation at 6328 Å. This is in good agreement with our frequency-domain NDFWM measurement.

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QMA7 Fig. 1. Apparatus for NDFWM spectroscopy. Scans of the oscillator controlling the probe modulator permit synthesized tuning of the optical frequency offset by steps as small as 10 mHz at visible wavelengths.



QMA7 Fig. 2. Signal-averaged spectrum showing the 1.8- and 372-Hz resonances. The full figure shows the high-resolution scan of the narrow central resonance with data points separated by 200-mHz intervals over a range of 10 Hz. The inset shows a 2000-Hz scan obtained with the novel intermodulation technique described in the text. The solid curves give the best fit Lorentzian line shape for each resonance.

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3:00 pm Invited

QThK1 Cooperative optical nonlinearities

S. C. Rand

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Arbor, Michigan 48109-2122*

Nonlinear-optical processes based on the localized response of individual atoms to strong applied optical fields are well known. In addition, strong nonlinearities have been demonstrated in recent years for delocalized excitations in organic molecules and artificial structures like quantum wells and superlattices. Here, however, we present observations and theory of a novel delocalized mechanism for nonlinear optics due to cooperative population dynamics in the excited states of rare-earth solids. Cooperative nonlinearities are shown to exhibit unusual features, such as threshold incident intensity, and are expected to exhibit temporal oscillations¹ and intrinsic chaotic behavior in strong coupling limits.²

Experiments have been performed at room temperature in a 2% Tm-doped crystal for which nonlinear absorption is observed on the 3H_4 - 1G_4 excited state absorption (ESA) transition (Fig. 1). Ground-state absorption at this wavelength is negligible. Nevertheless, increasing absorption appears owing to a sharp increase or "avalanche" in excited-state population, as verified directly by 3H_4 emission measurements at 1.9 μm versus intensity (Fig. 2).

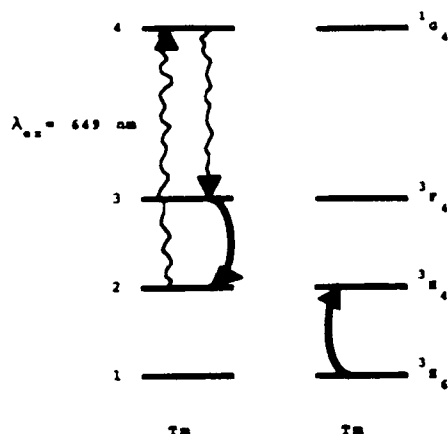
In highly doped crystals, Tm ion pairs can pool their energy by virtue of exchange or multipole coupling. A ground-state ion can be spontaneously promoted to the 3H_4 state as a 2F_4 ion decays to the same state in an energy-conserving, spontaneous cross-relaxation process. In this way the light induces a two-for-one process, and a sudden, substantial growth of excited-state population occurs. This effect has been exploited previously in other systems for the initiation of upconversion laser action.³ Here, however, we draw attention to the associated optical nonlinearity, which gives rise to strong four-wave mixing processes.

Novel two-beam coupling techniques were used to evaluate the third-order cooperative susceptibility as a function of intensity. The beam from a single-frequency dye laser was split into a strong pump beam and a weak transmitted probe beam whose in-phase intensity was recorded versus detuning. Programmable detuning of the probe beam was accomplished with acousto-optic modulation for continuous spectral scans over a range of 10 mHz-10 MHz. The results for n_2 versus intensity are shown in Fig. 3.

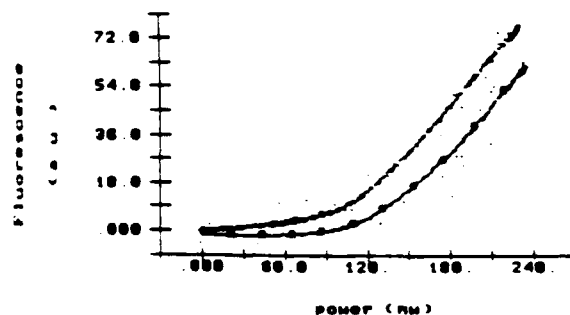
The key results are that the measure susceptibility shows an abrupt increase at the cooperative nonlinearity threshold and system response time increases. Continuous optical power of only a few tens of milliwatts with moderate focusing generates sizeable nonlinearities. Density matrix theory using pair states of the model in Fig. 1 also predicts population oscillations for interatomic coupling strengths (transfer frequencies), which exceed the natural decay rates of the ions, a condition believed to be satisfied on the rare-earth transition we have investigated. Hence, the nonlinear susceptibilities themselves should exhibit temporal oscillations, and calculations on a two-level model² suggest that chaotic fluctuations may also be encountered under appropriate driving conditions.

This research was supported by the Air Force Office of Scientific Research.

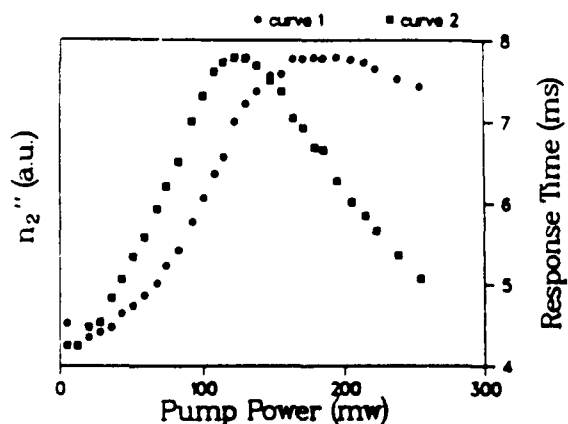
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QThK1 Fig. 1. Schematic illustration of cooperative dynamics of Tm ions. For each ion excited out of state 2 by incident light tuned to the ESA transition 2 to 4, two excited ions are obtained by way of a strong cross-relaxation process involving one state 3 ion and one ground-state ion. This leads to significant depopulation of the ground state and the appearance of increased absorption at the laser wavelength.



QThK1 Fig. 2. Upper curve: fluorescence intensity on the 3H_4 - 3H_6 transition at $1.9 \mu m$ versus incident light intensity at the 3H_4 - 1G_4 excited-state absorption resonance. Lower curve: upconversion fluorescence at $475.6 nm$ owing to the 1G_4 - 3H_6 transition.



QThK1 Fig. 3. Nonlinear refractive index (○) and system response time (□) versus input intensity, derived from two-beam coupling experiments performed on the 3H_4 - 1G_4 excited-state absorption transition of trivalent Thulium.

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CTuW20 Cooperative upconversion lasers

Ping Xie and Stephen C. Rand

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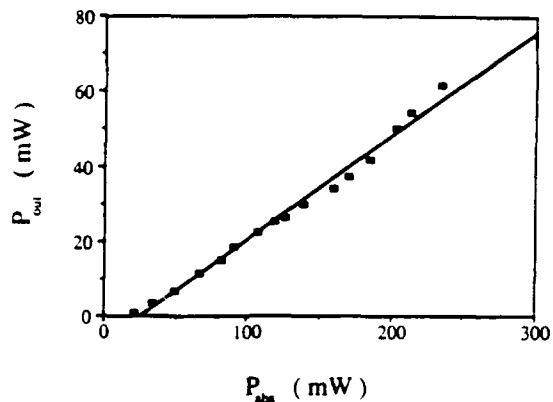
An important channel for upconversion relies on absorption of pump photons from excited states populated by pair upconversion. In previous¹ pulsed-Er upconversion lasers, this cubic contribution was small, because short excitation pulses were used, restricting ions from undergoing the slow first step of upconversion by this channel during the pulse-limiting upconversion to entirely multi-atom processes. However, cw excitation can maintain populations in upconverted states by pair processes, and direct absorption from such states may be important. Experiments reported here, however, establish that, in 5%Er: CaF₂ at 77 K, cw inversion is sustained by a cubic upconversion process primarily attributable to trio interactions. This work extends previous experiments² of cw lasing sustained by pair processes alone.

The experimental setup was similar to that used recently² for the pair-pumped erbium laser. Threshold for cw-laser operation on the self-terminating 0.85- μ m transition was reached for 10 mW of absorbed, incident light at 1.5 μ m in a monolithic cavity. A slope efficiency of 28% was measured, as indicated in Fig. 1. The mechanism of laser emission was investigated by measuring fluorescence intensity at 0.85 μ m as a function of excitation intensity and time. Results are shown in Figs. 2 and 3. The intensity dependence indicates cubic dependence on excitation, but does not distinguish between trio interactions and the alternative process involv-

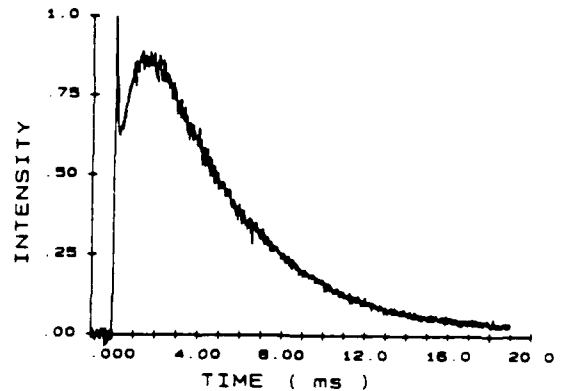
ing pair upconversion followed by absorption of a pump photon from the ⁴I_{9/2} state to reach ²H_{11/2}. We distinguished between these two possibilities by tailoring a pulse of 100- μ s duration acousto-optically from the pump beam at the upconversion laser threshold intensity and measuring time-resolved fluorescence at 0.85 μ m. A prompt and a long-lived component contribute to this fluorescence (Fig. 3). The prompt component has a rise time equal to the pulse-width and is evidently caused by the two-step, pair-mediated process. The slow component appears long after the excitation pulse is over, on a time scale characteristic of the directly pumped state ⁴I_{13/2}; in view of its cubic dependence on intensity this finding indicates trio upconversion.

A key result is that the integrated area of the trio component of the fluorescence decay in Fig. 3 is very much greater than the contribution from the competitive pair-mediated channel. Because 0.85- μ m fluorescence is a direct measure of population in the upper laser level and the ratio of these contributions remains constant as a function of pulsewidth at constant peak intensity, it is clear that trio contributions overwhelm pair-mediated contributions in the cw limit at these power levels.

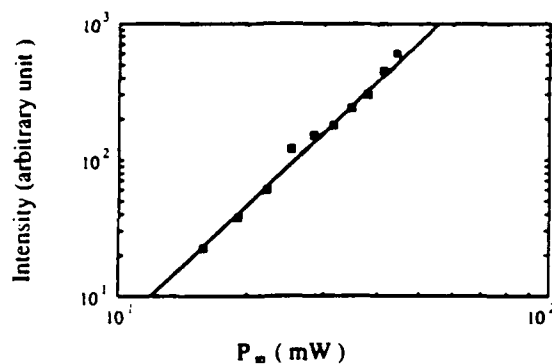
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CTuW20 Fig. 1. Cw laser output power at $0.85 \mu\text{m}$ as a function of absorbed input power at $1.5 \mu\text{m}$. Slope efficiency was 28%, and threshold power at $T = 77 \text{ K}$ was 10 mW.



CTuW20 Fig. 2. Fluorescence intensity from 5% Er: CaF_2 versus excitation intensity at a wavelength of $1.5 \mu\text{m}$. The solid curve has a slope of 3 to illustrate cubic dependence on excitation.



CTuW20 Fig. 3. Fluorescence intensity versus time at $0.85 \mu\text{m}$, following excitation at $1.5 \mu\text{m}$ with a $100\text{-}\mu\text{s}$ pulse having peak intensity fixed at the level of the upconversion laser threshold. The signal was heavily filtered optically and contained no contributions from other upconversion emission or scattered pump light.

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5:15 pm

CWL5 Beam coupling by cooperative nonlinearities

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Nondegenerate, two-beam coupling due to increasing absorption in Tm:YALO crystals at room temperature is reported. We measured the power-dependent nonlinear refractive index using a new technique. Also we explain the origin of this resonantly enhanced nonlinearity by cooperative dynamics involving pairs of coupled Tm ions in concentrated crystals.

We performed beam-coupling experiments with the apparatus of Fig. 1. Relative frequency detuning of the input beams was accomplished using acousto-optic modulators rather than Doppler shifting with moving mirrors. With this approach, the coupling between optical beams in saturable absorbers can be measured as a function of detuning in the range .01 Hz–100 MHz in a matter of seconds without signal processing. A unique aspect of the current experiment was that the laser was tuned to a region of transparency of the rare-earth-doped crystal. At a wavelength of 649 nm, strong upconversion fluorescence and beam coupling occurred above a minimum input light intensity. The spectrum observed in a single crystal of 2% Tm:YALO at room temperature is shown in Fig. 2, and exhibits the dispersion-shaped signal typical of nondegenerate, two-beam coupling in saturable absorbers.¹

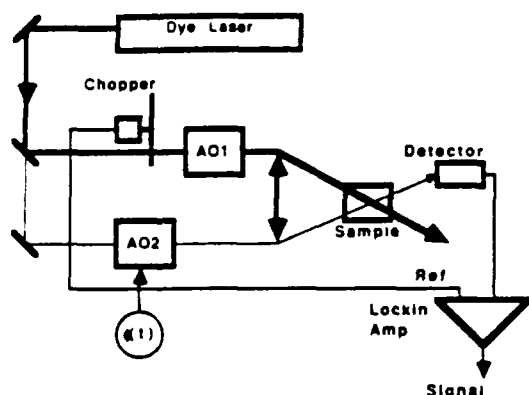
Using mode-mode coupling theory,² the two-beam coupling data was analyzed to obtain real and imaginary parts of the nonlinear refractive index and the system response time as a function of incident intensity. The results are shown

in Fig. 3 which revealed a rapid rise in the nonlinear refractive index at the onset of avalanche dynamics which occur³ in the 3H_4 state of Tm in this material. Also shown is the system response time which peaks at the avalanche threshold intensity.

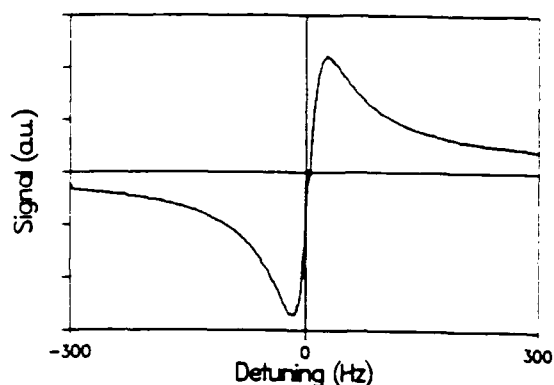
The absence of ground state absorption at 649 nm in Tm:YALO makes these results seem very surprising at first. However, at this wavelength the incident light is resonant with the excited state transition 3H_4 – 1G_4 and we have been able to show by direct measurement that a sudden increase occurs in the 3H_4 population on resonance at mid-range intensity in Fig. 3. This effect arises from coupling between an excited Tm ion and a ground state Tm ion in concentrated crystals, and is particularly strong in YALO presumably because of the unusually low non-radiative relaxation rate in this host. The coupling mediates cross relaxation which promotes enough ground state ions to the first excited state to induce total absorption at 649 nm in a crystal of only a few millimeters length, with only a few milliwatts of incident power.

Because the rate of cross relaxation clearly dominates natural decay rates above avalanche threshold, excited state ion-ion coupling is thought to be strong enough to delocalize the optical interaction. Hence, this new mechanism for beam coupling appears to be delocalized. Low power and room temperature operation make it an interesting candidate for many existing nonlinear optical applications.

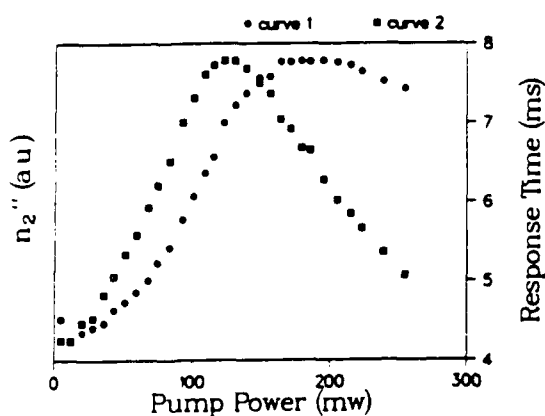
This research was supported by the Air Force Office of Scientific Research (AFSC).



CWL5 Fig. 1. Apparatus for two-beam coupling spectroscopy. Scans of the oscillator controlling the probe modulator permit synthesized tuning of the optical frequency offset by steps as small as 10 MHz at visible wavelengths.



CWL5 Fig. 2. Signal-averaged spectrum of two-beam coupling obtained by synthesized laser detuning technique. The entire spectrum is recorded during each scan and unlike previous methods, no signal processing is required.



CWL5 Fig. 3. Results for nonlinear refractive index (\circ) and system relaxation time (\square) as a function of incident intensity, which is derived from beam coupling data.

UPCONVERSION LASERS EXCITED BY PAIRS AND TRIOS

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Recent experiments in Er:YLF and other crystals have demonstrated efficient laser action from energy levels populated entirely by cooperative upconversion of pump energy from pairs and trios of excited Er ions.¹ This has revealed a general method for pumping solid state laser materials that is quite distinct from processes relying on the absorption of one or more photons by single ions. The new method permits excitation of high energy states by long wavelength radiation. For UV solid state lasers, it therefore offers an alternative to the use of deep ultraviolet pump sources that typically exhibit shallow penetration and can cause deleterious color center formation. Also, pair or multi-atom interactions can be exploited to achieve inversions on new transitions that may be normally self-quenching.

In 5% Er:YLF, pump energy at 1.54 μm promotes ground state Er ions to the first excited state where energy pooling takes place. A cooperative transition of excited, near-neighbor Er ions occurs subsequently that conserves energy overall and produces one high-energy ion and one or more ground state ions. Stimulated emission can in principle then occur between the upconverted energy state and lower levels at wavelengths as short as 0.8 μm for Er pairs or 0.5 μm for trios. To date, room temperature laser action from the upconverted states has been limited to a pair-pumped transition wavelength of 2.8 μm . But a noteworthy feature of this transition is that the lifetime of the lower laser level (23 ms) exceeds that of the upper level (4 ms), so that this transition is normally self-quenching. At cryogenic temperatures, trio-pumped laser action has been observed at wavelengths of 0.85, 1.23, and 1.73 μm .

The depletion rate of the pumped level, which is the lower laser level for the 2.8 and 0.85 μm emission lines, becomes a function of excitation in concentrated crystals, so that self-quenching can be avoided. True cw operation should be possible based on pair-pumping alone. Efficiencies achieved in the pulsed experiments have ranged so far

from 0.1–11.0% on four transitions despite a poor match between the Er:glass pump laser bandwidth and the ground state absorption spectra. Improved efficiencies and cw operation at shorter wavelengths are expected at lower temperatures using new pump sources.

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Continuous-wave, pair-pumped laser

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We report room-temperature operation of what we believe is the first continuous-wave laser that relies exclusively on cooperative upconversion by coupled ion pairs to achieve population inversion.

Conventional excitation of laser media raises individual atoms or ions directly to energy levels above the emitting level.¹ However, as we show here, energy states lying well above those populated by incident light can be reached by spontaneous, cooperative upconversion processes at rates sufficient to sustain stable, steady-state population inversions. Here we demonstrate cw laser action from upconverted states without significant contributions from multiphoton absorption. This novel operating principle holds promise as a means of pumping short-wavelength solid-state lasers without the need for radiation sources of still shorter wavelength, without the need for multiphoton absorption coincidences, and without the usual problems of color-center formation and short extinction length for pump light near the band edge of a solid.

While the cooperative upconversion pair process was previously found to account for modified kinetics in erbium lasers,² it has not been shown to be capable on its own of sustaining continuous inversion, as we show here. A variety of pulsed³ and cw upconversion^{4,5} lasers pumped by sequential multiphoton absorption processes at low temperatures have been reported. Also, earlier studies of upconversion lasers included those of pulsed operation of pair-pumped solid-state erbium lasers by Pollack *et al.*⁶ and Rand *et al.*⁷ In addition, conventional excitation was used to achieve cw operation on the 2.8- μm transition of erbium in CaF_2 (Ref. 6) and other hosts.⁸ Here, however, steady-state inversion of the upper laser level is achieved exclusively by upconversion due to spontaneous Er-Er interactions in the excited state. We also discuss why earlier studies^{2,6,7} or traditional laser analysis could not have predicted that true cw pair-pumped laser operation is even possible.

The energy-level diagram of the Er^{3+} ion is shown in Fig. 1. The crosses on level $^4I_{13/2}$ represent erbium ions initially prepared in this level by resonant absorption of light from a cw NaCl color-center laser operating at 1.51 μm . Pair interactions of near-neighbor ions can induce cooperative upconversion as shown in the figure, and similar processes have been the subject of research in gases since the work of Welch⁹ and in solids since the time of Dieke.¹⁰ The curved arrows indicate the dynamics schematically, with ion 2 losing energy by returning to the ground state and ion 1 gaining energy by upconversion to level $^4I_{9/2}$, which then relaxes rapidly [lifetime $\sim 7 \mu\text{sec}$ (Ref. 11)] to

populate the upper laser level $^4I_{11/2}$. In dilute $\text{Er}:\text{CaF}_2$ crystals the transition $^4I_{11/2} \rightarrow ^4I_{13/2}$ is self-quenched because the $^4I_{13/2}$ lifetime (28 msec) exceeds that of $^4I_{11/2}$ (13 msec) by roughly a factor of 2 at low excitation rates.⁶ However, Er-Er interactions in concentrated crystals make the $^4I_{13/2}$ lifetime excitation dependent. As a result, even when the lower laser level is pumped directly, as in the study reported here, the lower-state lifetime decreases with increasing occupation, and it becomes possible to achieve steady-state inversion from upconverted states without the assistance of multiphoton excitation.

This surprising result is in agreement with an exact analysis¹² in which the comparative rates of two-photon and two-atom pumping of the high-lying $^4I_{9/2}$ state may be varied. In our model each manifold is represented as a single level to furnish a simple but realistic picture of the basic dynamics. Within the rate-equation approximation, diagonal density matrix elements for the nonlinear four-level system are

$$\frac{d}{dt} \rho_{11} = \gamma_{41}\rho_{44} + \gamma_{31}\rho_{33} + \lambda_{21}\rho_{22} + \alpha\rho_{22}^2 - B_{12}I(\rho_{11} - \rho_{22}), \quad (1)$$

$$\frac{d}{dt} \rho_{22} = \gamma_{42}\rho_{44} + \gamma_{32}\rho_{33} - \gamma_{21}\rho_{22} - \kappa_{23}(\rho_{22} - \rho_{33}) - 2\alpha\rho_{22}^2 + B_{12}I(\rho_{11} - \rho_{22}), \quad (2)$$

$$\frac{d}{dt} \rho_{33} = \gamma_{43}\rho_{44} - \gamma_{33}\rho_{33} - \kappa_{23}(\rho_{33} - \rho_{22}), \quad (3)$$

$$\frac{d}{dt} \rho_{44} = \alpha\rho_{22}^2 - \gamma_{44}\rho_{44}, \quad (4)$$

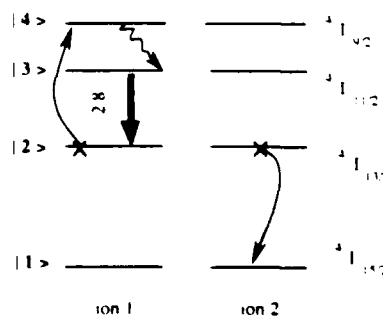


Fig. 1. Two-atom cooperative upconversion process responsible for steady-state inversion of level $^4I_{11/2}$ with respect to level $^4I_{13/2}$ in $\text{Er}:\text{CaF}_2$. The initial excited state, with both Er ions in the $^4I_{13/2}$ state, is prepared by cw irradiation at 1.51 μm on the $^4I_{15/2} \rightarrow ^4I_{13/2}$ absorption resonance.

where γ_{ij} is the spontaneous relaxation rate from level i to j , B_{ij} is the induced absorption rate, α is the pair upconversion coefficient, κ_{ij} is the rate of stimulated emission, and $\gamma_4 = \gamma_{43} + \gamma_{42} + \gamma_{41}$, $\gamma_3 = \gamma_{32} + \gamma_{31}$ are total decay rates of levels 4 and 3.

In Eq. (2) the term $-2\alpha\rho_{22}^2$ accounts for the loss of two ions simultaneously from level 2 during the cooperative upconversion process. One of these ions is destined for level 4 and one for level 1, giving rise to the positive α terms in Eqs. (1) and (4). This process populates levels higher than level 2. Steady-state solutions for the populations when two-photon absorption is turned off ($B_{24} = 0$) furnish the condition for population inversion between levels 2 and 3 ($\rho_{33}/\rho_{22} > 1$):

$$\alpha > \alpha_{th} \left[1 + \frac{\gamma_{43}(\gamma_{21} + \gamma_{11}) + \gamma_3(\gamma_4 + \gamma_{41})}{IB_{12}(3\gamma_{43} + \gamma_3)} \right]. \quad (5)$$

Here $\alpha_{th} = \gamma_3\gamma_4(3\gamma_{43} + \gamma_3)/N\gamma_{43}^2$ and $N = N_1 + N_2 + N_3 + N_4$. Inequality (5) shows that (ignoring cavity losses) steady-state inversion is possible for any non-zero pump intensity I , provided that the upconversion coefficient is sufficiently large.

For coupled-atom systems, however, the theoretical requirement for inversion given in inequality (5) does not guarantee stable cw operation in the same way as for conventional lasers. It is well known that, in oscillator systems with strong nonlinear coupling,¹³ unstable and even chaotic steady states are frequently encountered. Hence it is necessary to introduce small perturbations, linearize system equations about their steady-state values, and consider coherences to investigate stability of cw cooperatively pumped lasers.

We have performed such an analysis,¹² using the Routh-Hurwitz criterion for stability.¹⁴ The steady-state inversion due to Er pair interactions is predicted to be stable if γ_{43} is large compared with other decay rates in the system. According to our calculations, true cw operation (without chaotic behavior or sustained oscillations) should be achievable whenever the final state in the cooperative transition relaxes quickly, effectively preventing feedback through the reverse transition (cross relaxation). This result is relevant to our coupled-atom system in Er:CaF₂, where the $^4I_{9/2}$ lifetime is indeed short. In the opposite, coherent coupling limit, however, transient or sustained oscillations and population pulsations are expected to be prevalent in pair-pumped processes.

For the experiment, a crystal of 5% Er:CaF₂ was used as the pair-pumped laser medium. It consisted of a 3-mm-thick disk prepared with one flat surface and one convex surface of radius 2.5 cm. Both surfaces were antireflection coated in the range 1.4–1.6 μm , and between 2.7 and 2.9 μm the curved surface gave total reflection and the flat served as a 2% output coupler. The sample was pumped longitudinally at room temperature with a cw NaCl color-center laser focused by a 5-cm lens, absorbing 74% of radiation at 1.51 μm . For incident intensities above a threshold of 105 mW, TEM₀₀ laser emission was observed at 2.75 μm , as shown in Fig. 2. Cw output of 9 mW was obtained with 200-mW absorbed pump power, for an overall efficiency of 4.5%. Laser output was monitored through a notch filter at 2.8 μm (FWHM 10 nm),

using a fast InAs photodiode (rise time ~ 5 nsec) and a thermopile. Amplitude fluctuations at twice threshold were -15 dB below the dc level within the measurement bandwidth (dc 1 MHz).

Before the onset of saturation, the output power of the laser increased quadratically with incident power in excess of threshold. This behavior established the pumping mechanism as either two-photon absorption or pair-mediated upconversion. (Reabsorption of resonance fluorescence cannot produce population inversion on a self-terminating transition.) We distinguished between these two possibilities with the time-resolved fluorescence measurements described next.

The time dependence of the $^4I_{11/2}$ population was monitored directly by detecting fluorescence emission at 985 nm with a fast photomultiplier following short-pulse excitation of the laser medium. A rectangular pulse of 30- μsec duration and 100-mW peak power was selected acousto-optically from the cw pump beam for this purpose. Results are shown in Fig. 3(a).

The absorption of two pump photons must necessarily occur within the incident pulse duration. A calculation in which two-photon absorption accounts for 1%, 5%, or 40% of the upper-state population at short times results in the curves of Fig. 3(b), indicating that observed fluorescence would exhibit an obvious step by the end of the pulse in these cases. The measured rise of fluorescence, though, reveals different behavior. Instead, it shows virtually no prompt emission and appears over an interval consistent with the lifetime of the $^4I_{13/2}$ state in which pair interactions occur. These data immediately rule out contributions from two-photon absorption greater than approximately 1%, making it clear that long before the rate of two-atom cooperative upconversion reaches its peak, it is already 100 times more effective than two-photon absorption at the power levels of this experiment. A detailed fit of the temporal evolution in Fig. 3(a), followed by extrapolation to steady-state populations, reveals that two-photon excitation rate actually accounts for much less than 1% of the steady-state inversion.¹²

At present only a rough comparison of observed threshold intensity with that predicted from inequality (5) is possible. Using the values for decay rates given in Refs. 6 and 11, we find that $\alpha_t = 2.5 \times 10^{-19} \text{ cm}^3 \text{ sec}^{-1}$. Together with the only available estimate

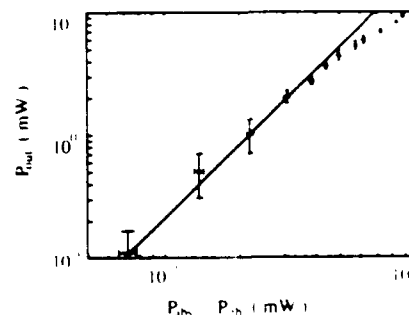


Fig. 2. Log-log plot of cw pair-pumped erbium laser output versus absorbed incident power above threshold. The solid curve has a slope of 2, illustrating the quadratic dependence followed at low powers. The onset of saturation is evident at high power.

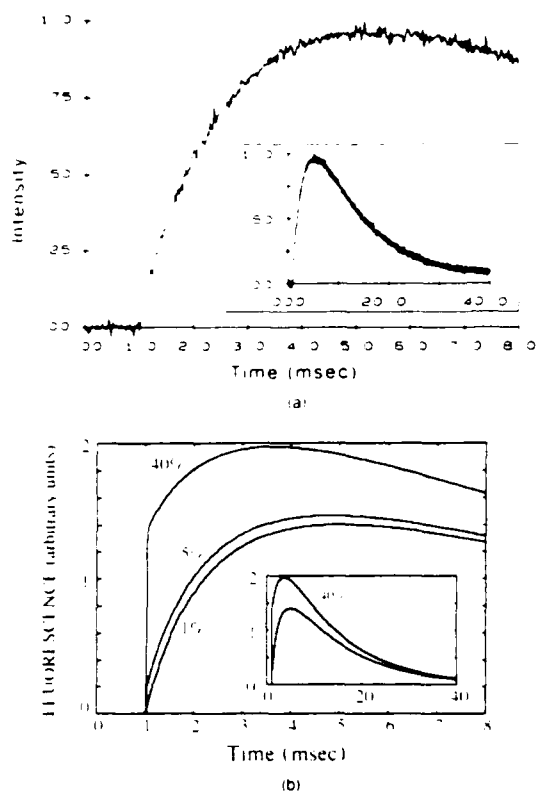


Fig. 3. (a) Fluorescence intensity at 985 nm versus time due to sample excitation at $t = 1$ msec by an acousto-optically tailored pulse of 30- μ sec duration. Peak intensity of the pulse (100 mW) equals the average power level used in cw excitation of the air-pumped laser. (b) Calculation of expected evolution of fluorescence when various proportions of excited-state population ρ_{33} are furnished by two-photon absorption during the 30- μ sec pulse interval.

of the upconversion coefficient, $\alpha = 0.32 \times 10^{-16} \text{ cm}^3 \text{ sec}^{-1}$,¹⁵ this yields $I_t = 2.1 \times 10^4 \text{ mW/cm}^2$. Considering that our α_t calculation ignores output coupling, and that incident power is not uniform over the nonlinear gain region, this estimate compares rather favorably with the observed threshold of $3.2 \times 10^5 \text{ mW/cm}^2$ for an estimated spot radius of 100 μm . Internal losses due to water absorption in the CaF_2 , pair upconversion from $^4I_{11/2}$, and higher-order upconversion processes have been ignored, and the true proportion of erbium impurities occurring in pair configurations is currently unknown. Contributions from these factors are expected to lead to increased estimates of I_t and to improved agreement with experiment.

In summary, we have demonstrated a cw laser pumped exclusively by cooperative upconversion from pairs of coupled atoms. Its detailed implementation as a solid-state erbium laser is remarkable in that cw operation is achieved on a self-quenched transition with lower-laser-level pumping. Its unoptimized efficiency is high enough for us to suggest that other cooperative pumping schemes should be of interest for short-wavelength solid-state lasers. Indeed, we recently operated a cw Er upconversion laser at 0.855 μm with 1.5- μm cooperative upconversion pumping, again on a self-terminating transition with lower-laser-level pumping. This general approach therefore offers the

prospect of new cw, rare-earth lasers in other highly doped crystals on both self-quenched and unquenched transitions.

Note added in proof: The excitation mechanism of the pulsed, pair-absorption-pumped laser reported in Ba/Tl vapor¹⁶ was quite distinct from that reported here. It relied on pair absorption on a collision-induced transition, a stimulated rather than a spontaneous process. Also, our result is fundamentally different from that of Quarles *et al.*,¹⁷ in which spontaneous cross relaxation (cooperative downconversion) of Tm pairs achieved inversion of Ho after an additional energy-transfer step.

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Continuous-wave trio upconversion laser

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We report operation of what we believe is the first continuous-wave laser which relies exclusively on cooperative upconversion by coupled ion trios to achieve population inversion.

The first laser to operate with an output wavelength shorter than that of the excitation was a pulsed erbium laser reported by Johnson and Guggenheim.¹ Since this early work, a variety of continuous-wave lasers have been reported in which upconversion occurs by different mechanisms, and inversions of upconverted energy levels have been sustained by avalanche processes,^{2,3} direct pair-pumping,⁴ and multiphoton absorption with variable contributions from energy transfer.^{5,6} To date, cw upconversion lasers have been operated in Pr-, Er-, Tm-, and Nd-doped solids, exhibiting surprisingly high efficiencies (up to 30%) in simple and robust, monolithic designs. All can potentially be pumped by diode lasers, and furnish wavelengths typically twice as short as diode lasers. With the advent of room-temperature operation these devices should therefore offer attractive alternatives to harmonic generation schemes⁷ for short-wavelength source applications.

In this letter we show that in addition to the known avalanche, two-atom and multiphoton upconversion mechanisms, steady-state inversions can also be maintained by cooperative transitions of atomic trios. Spontaneous cooperative fluorescence due to weakly coupled trios of rare-earth dopants in dielectric crystals has been studied previously.⁸ However, to our knowledge this is the first report of cw stimulated emission sustained by trio-pumping alone. Furthermore the 0.855 μm upconversion laser reported here achieves remarkably efficient cw oscillation on a self-terminating transition.

A simple, but realistic picture of the basic dynamics in the current work is obtained with a six-level model of trivalent erbium, indicated schematically in Fig. 1. Here each manifold is represented by a single level and cooperative transitions are permitted only in level two, which is considered to be very long lived at low excitation densities. In reality level three of Er^{3+} is also long lived, but its population arises entirely from level two pair processes. Hence it contributes to effective losses in the erbium pair laser⁴ and to quartic upconversion, but not to the cubic upconversion processes of interest here. In our model we draw on earlier results which justify omission of two-photon absorption source terms for level four population.⁴ However we include the possibility of direct absorption of pump photons by ions maintained in level four by cooperative pair upconversion to reach the upper laser level. Diagonal density matrix elements for this nonlinear system are

$$\frac{d}{dt}\rho_{11} = \gamma_{51}\rho_{55} + \gamma_{41}\rho_{44} + \gamma_{31}\rho_{33}$$

$$+ \gamma_{21}\rho_{22} + \alpha\rho_{22}^2 + 2\beta\rho_{22}^3 - B_{12}I(\rho_{11} - \rho_{22}) \quad (1)$$

$$\begin{aligned} \frac{d}{dt}\rho_{22} = & \gamma_{52}\rho_{55} + \gamma_{42}\rho_{44} + \gamma_{32}\rho_{33} - \gamma_{21}\rho_{22} + \kappa_{25}(\rho_{55} - \rho_{22}) \\ & - 2\alpha\rho_{22}^2 - 3\beta\rho_{22}^3 + B_{12}I(\rho_{11} - \rho_{22}) \end{aligned} \quad (2)$$

$$\frac{d}{dt}\rho_{33} = \gamma_{53}\rho_{55} + \gamma_{43}\rho_{44} - \gamma_{32}\rho_{33} \quad (3)$$

$$\frac{d}{dt}\rho_{44} = \gamma_{54}\rho_{55} - \gamma_{43}\rho_{44} + \alpha\rho_{22}^2 - B_{46}I(\rho_{44} - \rho_{66}) \quad (4)$$

$$\frac{d}{dt}\rho_{55} = \gamma_{65}\rho_{66} - \gamma_{54}\rho_{55} - \kappa_{25}(\rho_{55} - \rho_{22}) \quad (5)$$

$$\frac{d}{dt}\rho_{66} = -\gamma_{65}\rho_{66} + \beta\rho_{22}^3 + B_{46}I(\rho_{44} - \rho_{66}), \quad (6)$$

where the spontaneous relaxation rate between level i and j is given by γ_{ij} and B_{ij} is the induced rate between i and j . The pair and trio upconversion coefficients are α and β ,

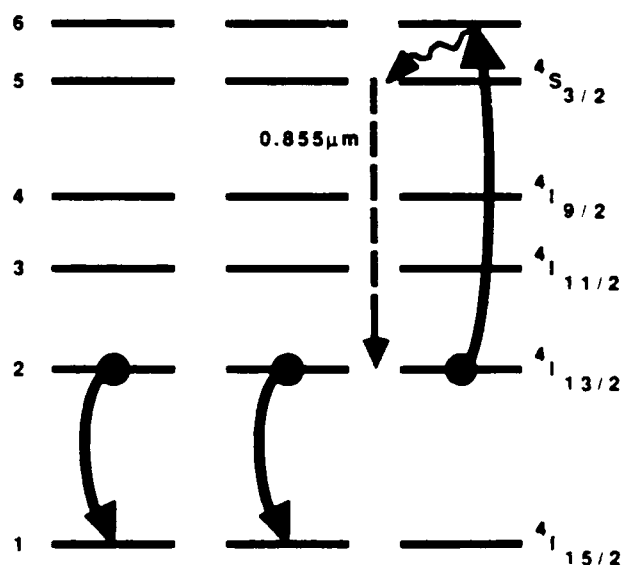


FIG. 1. Three-atom cooperative upconversion process responsible for steady-state inversion of level $4s_{3/2}$ with respect to level $4f_{15/2}$ in Er CaF_2 . The initial excited state, with three dopant ions in the $4f_{15/2}$ state, is prepared by cw irradiation at 1.51 μm on the $4f_{15/2} - 4f_{13/2}$ absorption resonance of trivalent erbium. The curved arrows indicate the dynamics schematically, with two atoms returning to the ground state while one is promoted to the upper laser level.

respectively. κ_{ij} is the rate of stimulated emission from i to j and γ_i is the total decay rate of level i .

In Eq. (2), there is a quadratic loss term corresponding to a pair process and a cubic term for trio upconversion. In the former process two ions are lost for each transition, requiring the indicated factor of 2. One ion returns to ground while the other is upconverted to level four. In the trio process three atoms leave level two, requiring a factor of 3 in the cubic loss coefficient. Two of the excited ions return to ground and one is upconverted directly to level six with subsequent rapid decay to level five. Of central importance here is the result that in the absence of cavity losses or pair upconversion a steady-state inversion between levels two and five can be maintained by trio upconversion alone if

$$B > \frac{\gamma_5(\gamma_5 + 3\gamma_6)^2}{\gamma_6^2} \left(1 + \frac{(\gamma_2 + \gamma_{51} + 2\gamma_5)\gamma_6}{(\gamma_5 + 3\gamma_6)B_{12}I} \right)^2. \quad (7)$$

Conditions such as (7) for steady-state inversion in coupled-atom systems do not guarantee stable cw operation. The oscillator system is highly nonlinear and may exhibit unstable or even chaotic states. Hence a stability analysis is required to ensure theoretically stable operation of the laser. We have analyzed trio laser stability with a simplified four-level model (omitting levels three and four) by introducing small perturbations and linearizing system response near steady-state conditions. The Routh-Hurwitz criterion⁹ then predicts stable oscillation whenever the highest state, the trio-pumped state, is short-lived compared to other levels in the system. This condition is well met in Er^{3+} .

To demonstrate an erbium trio laser experimentally, a 3-mm-thick crystal of 5% $\text{Er}:\text{CaF}_2$ was prepared with one flat surface and one convex surface of radius 2.5 cm. Both surfaces were antireflection coated in the range 1.4–1.6 μm . Additionally, in the range 0.8–0.9 μm , the curved surface was coated for total reflection ($R > 99.9\%$) and the flat served as a 0.5% output coupler. The sample was pumped longitudinally at liquid-nitrogen temperature with a cw NaCl color center laser focused by a 5 cm lens, and it absorbed 74% of incident light at 1.51 μm . For incident intensities above a threshold of 10 mW, TEM_{00} laser emission was observed at 0.855 μm as shown in Fig. 2. A maximum of 64 mW cw output was obtained for 235 mW of absorbed pump power, for an overall efficiency of 26% (theoretical maximum is 60%) and a slope efficiency of 28%. No evidence of saturation spiking behavior was observed and amplitude fluctuations were 15 dB below the output level within the measurement bandwidth (dc 1 MHz).

The intensity dependence of upconversion fluorescence shown in Fig. 3 reveals that level five is populated by a process varying with the cube of the incident intensity. This is a key result of the present work, since with our method of excitation, there are only two possible channels for populating upper laser level five by a cubic process. One channel is a trio process and the other is absorption of a pump photon by pair-pumped ions in level four. Additional conceptual possibilities related to ground state

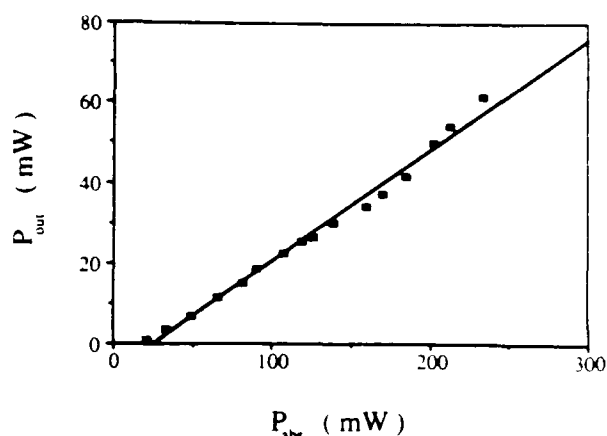


FIG. 2. Linear plot of cw trio-pumped laser output vs absorbed incident power. The solid curve is a linear guide to the eye.

two- or three-photon absorption are ruled out by the absence of prompt fluorescence from levels three and four, as discussed in our earlier paper.⁴ We distinguished between the two remaining possibilities with time-resolved fluorescence measurements shown in the next figure.

Figure 4 shows the time dependence of fluorescence at 0.855 μm from the upper laser level, monitored with a fast photomultiplier following pulsed excitation below threshold. A rectangular pulse of 100 μs duration and 8 mW peak power was selected acousto-optically from the cw pump beam for this purpose. Signal averaging of 4000 scans was used to improve signal-to-noise ratio on a time scale of milliseconds with 1 MHz bandwidth.

Two components are evident in the experimental curve. The first is a prompt component with a rise time equal to the pulse duration, followed by a subsequent, rapid decay. This component can only be due to pump absorption by pair-upconverted ions in state $^4I_{9/2}$, since the signal decays when the pulse ends and yet two-photon absorption contributions to the $^4I_{9/2}$ population are insignificant under these conditions (see Fig. 4 inset and Ref. 4). This component therefore corresponds to the pair-mediated channel. The second component rises slowly,

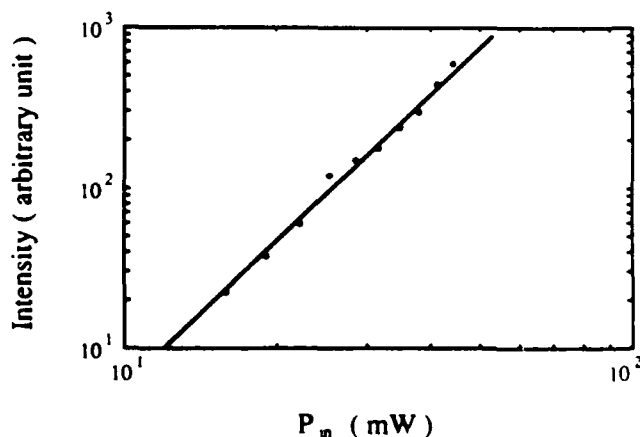


FIG. 3. Log-log plot of level five fluorescence vs excitation intensity. The solid curve illustrates a cubic dependence for 0.855 μm upconversion emission.

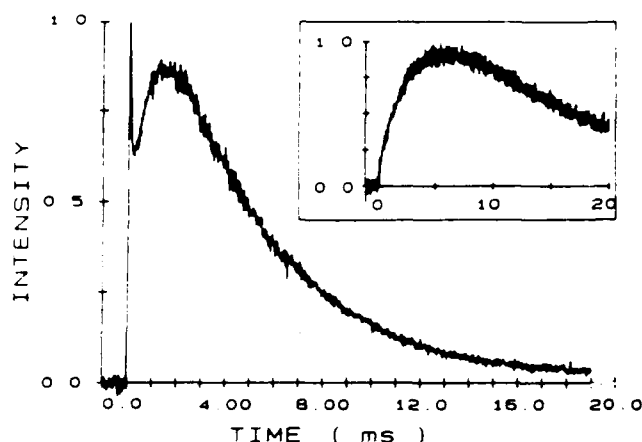


FIG. 4. Fluorescence intensity at 855 nm vs time, due to sample excitation by an acousto-optically tailored pulse of 100 μ s duration and 8 mW peak intensity. Prompt emission is observed as a spike during the applied pulse. A slowly rising component appears long after the pulse terminates. The relative importance of these two excitation channels can be assessed by comparing areas under component curves. The inset shows the absence of prompt emission at 0.98 μ m, ruling out level four two-photon and level six three-photon excitation.

reaching its maximum long after the excitation pulse is over. The only remaining process capable of furnishing cubic upconversion to $^4S_{3/2}$ is one involving three excited $^4I_{13/2}$ ions. Hence the second component corresponds to this trio contribution. Notice that long before the rate of trio upconversion reaches its peak in the steady state its contribution to upper laser level population exceeds the pair-mediated portion by a factor of 40, estimated from relative areas under the curve.

The overall efficiency of this trio-pumped laser is 26% at 235 mW pump power. Its slope efficiency is nearly 30%, using a linear approximation for the output curve versus input, and as mentioned earlier its threshold is only 10

mW. This trio laser consequently has much higher efficiency and lower threshold than the pair-pumped erbium laser,⁴ a somewhat unexpected result due at least partly to the absence of water absorption at the emission wavelength as well as higher cavity Q . However the cubic depletion rate of the lower level may also play a significant role. In addition, amplitude fluctuations are much smaller in trio laser output, a result possibly due to the very short lifetime of level six which discourages back transfer.

In summary, we have demonstrated a continuous-wave laser pumped by cooperative upconversion from trios of excited, coupled erbium ions in CaF_2 . Its unoptimized efficiency is high, indicating that trio mechanisms by themselves can be exploited for the development of novel, short-wavelength solid-state lasers. The erbium trio laser is remarkable in that cw operation is achieved on a self-terminating transition with lower laser level pumping. Hence this nonlinear pumping scheme not only offers the prospect of new upconversion lasers, but also of cw rare-earth lasers in other highly doped crystals on previously unusable, self-terminating transitions.

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Avalanche upconversion in Tm:YALO₃

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We report the sudden appearance of upconversion fluorescence and resonant increasing absorption above a threshold intensity in Tm:YALO₃ at room temperature. These effects are shown to arise from nonlinear, cooperative, excited-state dynamics. Comparison with quantum theory reveals essential roles both for pairwise cross relaxation and excitation migration among Tm impurity ions.

Absorption features with thresholds in rare-earth crystals were first observed in Pr:LaCl₃ and Pr:LaBr₃ by Chivian *et al.*¹ in spectral regions devoid of ground-state transitions. Their experiments were a prelude to development of avalanche upconversion lasers^{2,3} operating at shorter wavelengths than their excitation and sustained by optical pumping in regions of transparency. While empirical modeling of the nonlinear absorption⁴ observed under avalanche conditions has supported a picture that involves cooperative energy-pooling pair processes between excited and neighboring ground-state ions, basic premises of this model have not been directly tested experimentally. Furthermore no microscopic theory has been advanced that explains observations made to date consistently.

In this Letter we present experimental results on avalanche absorption and upconversion in a new system at room temperature as well as a quantum theoretical framework for calculating system behavior. We show that when incident light is tuned away from ground-state absorption wavelengths and into resonance with an excited-state transition of Tm, nonlinear absorption and upconversion fluorescence appear above a distinct threshold intensity. We verify directly that population in the lower resonant excited state undergoes a rapid increase that may aptly be called an avalanche and cannot be explained by linear dynamics. Theoretical analysis indicates that the cooperative dynamics responsible for the avalanche involve two distinct processes, namely, near-neighbor cross relaxation and excitation migration between pairs. Collectively, these results furnish a consistent conceptual basis for a detailed understanding of avalanche upconversion and show that in selected media this phenomenon can occur at high enough temperatures to render applications practical.

Figure 1 displays both the low-intensity absorption spectrum and the excitation spectrum of induced absorption features near 649.5 nm in the biaxial host crystal YALO₃. A continuous-wave, single-mode DCM ring dye laser polarized along a principal axis was tuned to the wavelength of the excited-state transition of Tm³⁺ at 649.5 nm (Fig. 2) and focused to a spot radius of 45.0 μ m in an 8-mm-thick sample. The dopant concentration was 2%.

Absorption between 640 and 660 nm was negligible at low incident intensities. However, as shown in Fig. 3, transmission in Tm:YALO₃ decreases sharply at a resonant wavelength of 649.5 nm above intensities of 9.4×10^2 W/cm². This wavelength corresponds precisely to the wavelength of the ³H₄(1)–¹G₄(1) excited-state transition of Tm.⁵ Absorption becomes so highly nonlinear above threshold that an input beam of Gaussian profile evolves into a doughnut distribution with nearly zero transmission at beam center and relatively high transmission in the wings. At the same time strong blue upconversion fluorescence is emitted by the crystal.

We investigated the mechanism responsible for this nonlinear absorption and upconversion by first noting that efficient cross relaxation between levels ³F₄ and ³H₆ of Tm occurs in a wide variety of crystals doped heavily with this ion.⁶ This process is illustrated in Fig. 2, and, since the lower level (³H₄) of the induced absorption transition is the same as the intermediate state in cross relaxation, it is natural to inquire about its potential role in avalanche dynamics. To investigate excited-state dynamics, we recorded fluorescence at two wavelengths as a function of incident intensity with the laser tuned to the strongest excited-state resonance. Long-wavelength fluorescence at 1.9 μ m [³H₄(1)–³H₆(1,2)] was detected with an InAs photodiode and used to monitor ³H₄ excited-state population directly. Short-wavelength emission at 475.6 nm [¹G₄(1)–³H₆(1)] was used to monitor upconversion behavior versus intensity at 649.5 nm.

Fluorescence intensity is strictly proportional to the initial-state population, irrespective of the final-state dynamics. Hence results for emission at 1.9 μ m versus the incident intensity (Fig. 4) constitute a direct measurement of population in the ³H₄ excited state. Absorption at 649.5 nm, on the other hand, reflects population differences of the ³H₄ and ¹G₄ levels rather than of the avalanche population itself. Short-wavelength emission at 475.6 nm again samples population in the upper resonant state directly. Our results clearly show that the population itself in the ³H₄(1) level of Tm exhibits a dramatic increase above a relatively sharp threshold intensity. At the same time, increasing absorption

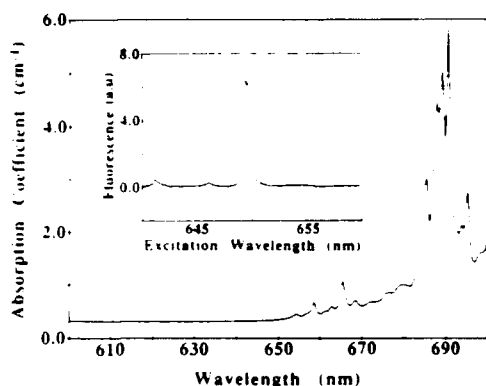


Fig. 1. Unpolarized absorption spectrum of 2% Tm:YAlO₃ at room temperature and low intensity. Induced absorption resonances appear between 640 and 650 nm at intensities above 10³ W/cm², as shown by the inset excitation spectrum of 475.6-nm fluorescence. In the inset, the main resonance is due to the ³H₄(1)–¹G₄(1) excited-state transition of Tm³⁺. Smaller features at 646 and 641 nm correspond to the ¹G₄(2)–³H₄(1) and ¹G₄(4)–³H₄(1) avalanche transitions, respectively.

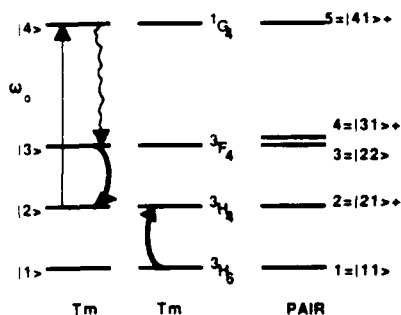


Fig. 2. Left: energy-level diagram illustrating cooperative relaxation of neighboring Tm ions. The straight arrow indicates an absorptive optical transition at frequency ω_0 followed by intraconfigurational relaxation (wiggly arrow). The curved arrows indicate the subsequent spontaneous pair process in which one ³F₄ ion relaxes to ³H₄, while its neighbor is promoted to the same state in a nearly resonant cooperative transition. Right: levels of the pair basis formed from uncoupled product states. States labeled + indicate that for degenerate states only the symmetric Davydov component is presumed to participate in dynamics, having nonzero matrix elements for optical transitions to ground and other + states in the presence of inversion symmetry of the pair [$|21\rangle+ = 2^{1/2}(|2\rangle|1\rangle+|1\rangle|2\rangle)$].

and upconversion fluorescence appear with the same threshold, in qualitative agreement with earlier observations on Pr ions.^{1,4} Here, however, the avalanche level population is monitored directly rather than indirectly, no abrupt jumps in system parameters are observed, and room-temperature operation is achieved. Additionally, theoretical analysis of these data, which we turn to now, achieves good agreement with experiment and reveals new aspects of the avalanche upconversion process not recognized previously.

Descriptions of avalanche dynamics to date have focused on pair interactions using rate equations. However, the rate approximation provides an incomplete theory for the avalanche effect for two reasons.

First, with this approach, cooperative population decay rates (probabilities) of neighboring atoms are written in terms of uncorrelated products of occupation probabilities for individual atoms. This is contrary to the basic picture of the cooperative mechanism responsible for the avalanche in which neighboring ions make simultaneous (and therefore completely correlated) energy-conserving transitions mediated by short-range interactions.¹ Second, cooperative relaxation rates must dominate natural decay rates when avalanche effects are observed, so that the implicitly perturbative approach of rate equations that ignores coherences is not justified *a priori*. To include interion coherences, we started with the Liouville equation,

$$i\hbar \frac{d}{dt} \rho = [H, \rho] - i\hbar \Gamma(\rho)\rho, \quad (1)$$

where $H = H_0 + H_{\text{int}} + V$ is the Hamiltonian, H_{int} is the interion coupling (multipole or exchange interac-

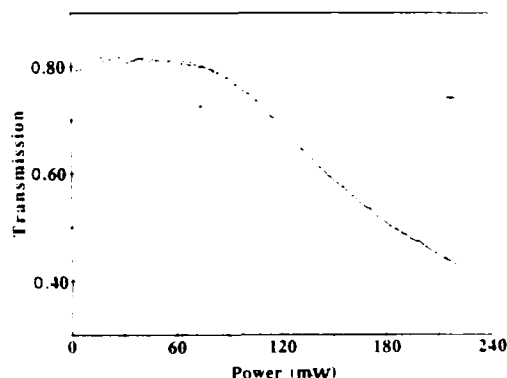


Fig. 3. Total Gaussian beam transmission through 2% Tm:YAlO₃ at 649.5 nm as a function of intensity ($T = 293$ K).

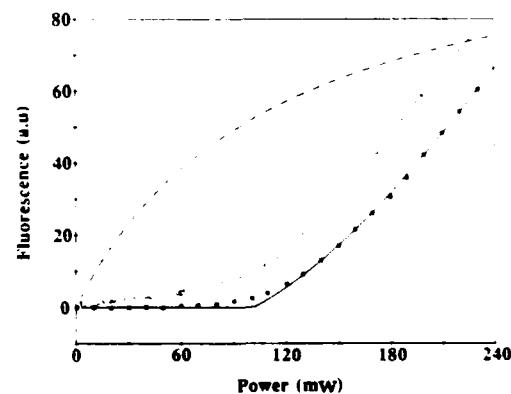


Fig. 4. Theoretical and experimental excited-state populations in 2% Tm:YAlO₃, excited with monochromatic light at 649.5 nm. Asterisk and dotted experimental traces: upconversion and infrared fluorescence at 475.6 nm and 1.9 μm , respectively. Dashed curve: calculation of ¹G₄ population in isolated pairs ($\alpha = 0$), indicative of increasing ³H₄ population (magnified by 2×10^{11} to be visible on the scale of other calculations). Solid curve: numerical ¹G₄ results combining intrapair relaxation with energy migration between distant pairs (best fit $\alpha = 2.0 \times 10^6 \text{ s}^{-1}$). The fixed parameters are $\lambda = 2.8 \times 10^{-10} \text{ s}^{-1}$, $|H_{34}|^2/\hbar^2\Gamma_{34} = 10^3 \text{ s}^{-1}$, $\gamma_{31} = \gamma_{42} = \gamma_{43} = \gamma_{51} = \gamma_{52} = \gamma_{53} = 0$, $\gamma_3 = \gamma_{32} = 107 \text{ s}^{-1}$, $\gamma_4 = \gamma_{41} = 780 \text{ s}^{-1}$, and $\gamma_5 = \gamma_{54} = 3.6 \times 10^3 \text{ s}^{-1}$.

tions), and V describes the light-matter interaction. The relaxation rate given by phenomenological damping term $-i\hbar\Gamma(\rho)\rho$ in our semiclassical approach is linear in atomic density for intrapair dynamics in a pair basis of states but bilinear for relaxation between uncorrelated pairs. Γ depends on ρ since decay can occur not only internally but also by random hopping of excitation to a distant pair with a rate dependent on pair populations at the origin and destination. Equation (1) in the uncoupled pair basis of Fig. 2 yields the following equations for populations in pair levels coupled by the optical field:

$$\frac{d}{dt}\rho_{22} = (V_{25}\rho_{52} - \rho_{25}V_{52})/i\hbar - \gamma_{21}\rho_{22} + \lambda(\rho_{11} - \rho_{22}) + \gamma_{32}\rho_{33} + \gamma_{42}\rho_{44} + \gamma_{52}\rho_{55} + 2\alpha\rho_{11}\rho_{33} - 2\alpha\rho_{22}^2, \quad (2)$$

$$\frac{d}{dt}\rho_{55} = (V_{52}\rho_{25} - \rho_{52}V_{25})/i\hbar - \gamma_{51}\rho_{55}. \quad (3)$$

These equations must be solved self-consistently with those for populations in other levels, as well as for optical and interatomic coherences,

$$\frac{d}{dt}\rho_{25} = i\omega_0\rho_{25} + V_{25}(\rho_{55} - \rho_{22})/i\hbar - \Gamma_{25}\rho_{25}, \quad (4)$$

$$\frac{d}{dt}\rho_{34} = \rho_{34}(H_{33} - H_{44})/i\hbar + H_{34}(\rho_{44} - \rho_{33})/i\hbar - \Gamma_{34}\rho_{34}, \quad (5)$$

These equations reduce to rate equations when the interatomic coherence is zero ($H_{34} = \langle 3|H_{\text{int}}|4 \rangle = 0$). γ_{ij} is the decay rate from level i to j , γ_k is the total rate from level k , and ρ_{kl} is the density-matrix element between pair levels k and l . Γ_{ij} is the rate of relaxation of coherence between levels i and j , and λ is a thermal pumping rate, assumed to be effective solely in populating the first excited pair state. α is the rate of resonant energy migration between distant pairs (ρ is dimensionless).

Coefficient α accompanies nonlinear terms that describe dynamics between uncorrelated pairs. For example, the term $+2\alpha\rho_{11}\rho_{33}$ in Eq. (2) describes the relaxation of two pairs initially in pair states 3 and 1. Energy exchange yields two pairs in state 2 at a rate α . Linearized theory ($\alpha = 0$) describes coherent dynamics within isolated pairs and predicts a minute amount of increasing absorption, as indicated by the dashed curve in Fig. 4. However, it does not reproduce other experimental observables such as the threshold of avalanche absorption, its qualitative dependence on intensity, or its magnitude. Hence cooperative decay within isolated pairs is by itself inadequate to account for avalanche behavior.

Individual pairs undergoing cross relaxation can increase the population of the avalanche level but can at most double the weak initial absorption due to overlapping ground-state transitions or thermally excited Tm ions in the first excited state. Hence isolated pairs can initiate minor increases but cannot account for strong induced absorption. Correlated dynamics within isolated pairs is therefore

an essential mechanism by which ions reach an ordinarily empty excited state, but a mechanism must also exist for rapid excitation migration involving more ions to achieve substantial increases in absorption.

Evidence for this is presented in Fig. 4, in which the solid curve shows the result when α is taken to be nonzero. This introduces resonant energy migration (random hopping) between state-3 pairs and distant ground-state neighbors, which constitute a reservoir of excitable atoms. For simplicity, we disregard the detailed nature and distance dependence of the migration process ($\alpha = \text{constant}$), retaining only a bilinear dependence on occupation probabilities of pairs at the origin and destination in the corresponding relaxation rate.

Good agreement with experiment is obtained with the full theory, which permits a majority of impurity ions to participate in cooperative dynamics through combined intrapair and interpair relaxation. The small discrepancy in Fig. 4 between the solid theoretical curve and data near threshold is attributed to nonuniformity of interaction across the Gaussian beam. Agreement is therefore only obtained when the internal dynamics of near-neighbor atomic interactions is followed by energy transfer to more distant Tm ions, with a transfer rate independent of the identity of the initial pair by virtue of randomness in the migration process. Clearly, slow near-neighbor dynamics yield increasing absorption, whereas fast migration yields threshold behavior and immensely magnifies the excited-state absorption.

In summary, we have identified two important aspects of avalanche dynamics, describable by a single set of equations. First, light that is not resonant with any ground-state absorption promotes ions to excited states by activating short-range interactions between one excited ion and a near neighbor in the ground state, which generates increased absorption. Second, energy migration multiplies the effectiveness of the cooperative dynamics, which introduces a characteristic absorption threshold. In 2% Tm:YALO, avalanche absorption and upconversion are observed at room temperature and are well described by density-matrix theory.

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